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BY:

Date: October 3, 2005

MAIL STOP AMENDMENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Patent Application of: :
Atsushi Ueda et al. :
Conf. No.: 7373 : Group Art Unit: 1745
Appln. No.: 10/058,707 : Examiner: Raymond Alejandro
Filing Date: January 28, 2002 : Attorney Docket No.: 10059-404US
Title: Non-Aqueous Electrolyte Secondary Battery (P27007-01)

FOURTH DECLARATION OF ATSUSHI UEDA UNDER 37 C.F.R. § 1.132

I, Atsushi Ueda, declare and state as follows:

1. I am a co-inventor of the invention described and claimed in the above-identified patent application. I am the same Atsushi Ueda who executed first, second and third Declarations under 37 C.F.R. § 1.132, filed in this application on June 15, 2004, November 8, 2004, and June 23, 2005, respectively.

2. I am familiar with the above-referenced application, and in particular with the Office Action dated July 6, 2005 (Paper No. 20050629). I am submitting the present Declaration to overcome the obviousness-type double patenting and § 103(a) rejections of claims 1-6 and 8-15 by demonstrating the effects observed on the amount of gas generated after cycles of a secondary battery when the non-aqueous solvent contains 0.5 to 20 vol% cyclic carbonic acid ester (B).

The Examiner's Position and the Purpose of the Declaration

3. The Examiner has taken the position that the closest prior art to the invention includes U.S. Patent No. 6,723,473 ("the '473 patent"); U.S. Patent Application Publication No. 2003/0118913 of Takami et al. ("Takami"), WO 01/03228 ("WO '228"), EP 0 796 510 ("EP '510"), JP 08-96852 ("JP '852"), U.S. Patent Application Publication No. 2001/0018152 of Kida et al. ("Kida"), U.S. Patent No. 6,090,506 of Inoue et al. ("Inoue"), and U.S. Patent Application Publication No. 2002/0001756 of Hamamoto et al. ("Hamamoto").

4. In the Office Action dated July 6, 2005, the Examiner rejected claims 1-6 and 8-15 under 35 U.S.C. § 103(a) as being obvious over combinations of Takami, JP '852 and WO '228 in view of EP '510, Kida, Inoue and/or Hamamoto. The Examiner argued that all of the claimed elements are taught or suggested by the proposed combinations of cited references, including the inclusion of VC and VEC in the claimed volume range in a solvent mixture containing a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C).

5. The purpose of this Declaration is to demonstrate that when 0.5 to 20 volume percent of a combination of VEC and VC is included in a solvent mixture containing components (A) and (C), a dramatic decrease in the amount of gas generated after cycles is observed relative to the amount of gas generated when greater or lesser amounts of VEC and VC are included in the solvent mixture with components (A) and (C). These results would not be expected based on the prior art, which does not teach or suggest a solvent containing both VEC and VC nor the preferred ranges thereof, and such a demonstration will thus overcome the rejections of claims 1-6 and 8-15.

Background and Purpose of the Invention

6. The non-aqueous electrolyte secondary batteries according to the present invention were developed to overcome problems known in the art, such as deterioration of charge and discharge characteristics. The batteries according to the invention have excellent charge and discharge characteristics, particularly at low temperature, and show satisfactory charge and discharge characteristics, even after being exposed to high temperatures over a period of time.

7. According to the presently claimed invention of claims 1-6 and 8-15, non-aqueous electrolyte secondary batteries which exemplify these properties are realized by using a non-

aqueous solvent containing: (A) a cyclic carboxylic acid ester; 0.5 to 20 volume % of (B) a cyclic carbonic acid ester having at least one carbon-carbon unsaturated bond and comprising vinylethylene carbonate (VEC) and vinylene carbonate (VC); and (C) a cyclic carbonic acid ester having no carbon-carbon unsaturated bond. Cyclic carboxylic acid esters (A) are prone to decomposition by hydrofluoric acid, possibly deteriorating the cycle characteristics of the battery. The use of LiBF₄ also deteriorates cycle characteristics of the battery because the degree of electrolytic dissociation of LiBF₄ is small or a film formed on the surface of the negative electrode comprising graphite is weak. However, as shown in Tables 10 and 11 of the present application, the use of both salts in combination, particularly in the claimed ratio, improves cycle characteristics of the batteries and decreases the amount of gas generated, and also results in favorable capacity maintenance rates and cycle life. Further, the use of components (C) and VEC in combination with component (A) results in favorable battery characteristics which overcome these disadvantages. The use of VC and VEC in combination is particularly desirable because VEC suppresses the reaction activity of VC at high temperatures to improve storage characteristics of the battery at such temperatures.

8. In order to demonstrate the effect of including 0.5 to 20 vol% of a cyclic carbonic acid ester (B) containing both VEC and VC in the non-aqueous solvent on the amount of gas generated after cycles, a series of batteries was prepared using different concentrations of component (B), and the amounts of gas generated after cycles for each of these batteries were measured and compared.

Experimental Procedure

9. Batteries were produced using the procedure described in Example 6 of the above identified application. Non-aqueous electrolytes were prepared by dissolving LiBF₄ in each of the solvent mixtures shown in Table 1 at a LiBF₄ concentration of 1 mol/L. In all cases, the ratio of VC to VEC was 1:1.

Table 1

Battery	cyclic carboxylic acid ester (A) (vol %)	cyclic carbonic acid ester (B) (vol %)	cyclic carbonic acid ester (C) (vol %)	solute	solute concentration (mol/L)		
	GBL	VC	VEC	VC + VEC	EC		
1	79.9	0.05	0.05	0.1	20	LiBF ₄	1
2	79.5	0.25	0.25	0.5	20	LiBF ₄	1
3	79	0.5	0.5	1	20	LiBF ₄	1
4	75	2.5	2.5	5	20	LiBF ₄	1
5	70	5	5	10	20	LiBF ₄	1
6	60	10	10	20	20	LiBF ₄	1
7	50	15	15	30	20	LiBF ₄	1

(EC= ethylene carbonate; VEC = vinylethylene carbonate; GBL = gamma-butyrolactone; VC = vinylene carbonate)

10. As in Example 6 of the application, each of the seven batteries in Table 1 above was charged at a constant current and constant voltage at an ambient temperature of 20°C, a fixed ceiling voltage of 4.2 V, and a maximum current of 1050 mA for 2.5 hours. The charged batteries were then discharged at a discharge current of 1500 mA at 20°C until the voltage reached 3.0 V. The amounts of gas generated in the batteries after the cycles were then measured.

Results and Discussion

11. Table 2 shows the results obtained by measuring the amounts of gas generated after cycles for each of the seven batteries of Table 1.

Table 2

Battery	vol. % component (B)	Amount of Gas Generated After Cycles (mL)
1	0.1	4.9
2	0.5	3.6
3	1	2.7
4	5	2.1
5	10	2.6
6	20	3.3
7	30	4.8

12. As shown in Table 2, when the amount of cyclic carbonic acid ester (B) was 0.1 vol % (battery 1) or 30 vol % (battery 7), the amount of gas produced was as high as 4.9 ml or 4.8 ml, respectively. However, when the concentration of component (B) was 0.5 to 20 vol % as claimed (batteries 2-6), the amounts of gas produced were reduced to no more than 3.6 ml, and were as low as 2.1 ml. This demonstrates that when the concentration of cyclic carbonic acid ester (B) in the non-aqueous electrolyte is limited to 0.5 to 20 vol%, the amount of gas produced after cycles can be significantly reduced.

Conclusion

13. This Declaration demonstrates that by preparing non-aqueous electrolyte secondary batteries according to the claimed invention, which contain 0.5 to 20 vol % VC and VEC in combination with a cyclic carboxylic acid ester (A) and a cyclic carbonic acid ester (C) having no carbon-carbon unsaturated bond, favorable and unexpected results are obtained, particularly with regard to amount of gas generated after cycles. None of the prior art of record specifically teaches the use of these particular concentrations of VC and VEC in combination with components (A) and (C) in the non-aqueous solvent, nor makes obvious the results which have been observed by the present invention.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that those statements were made with the knowledge that willful false statements the like so made are punishable by fine or imprisonment, or both, under Section 1003 of Title 18 of the United States Code; and that such willful statements may jeopardize the validity of the application or any patent issuing thereon.

Dated: September 27, 2005

Atsushi Ueda

Atsushi Ueda